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Author: Scott F. DeMuth

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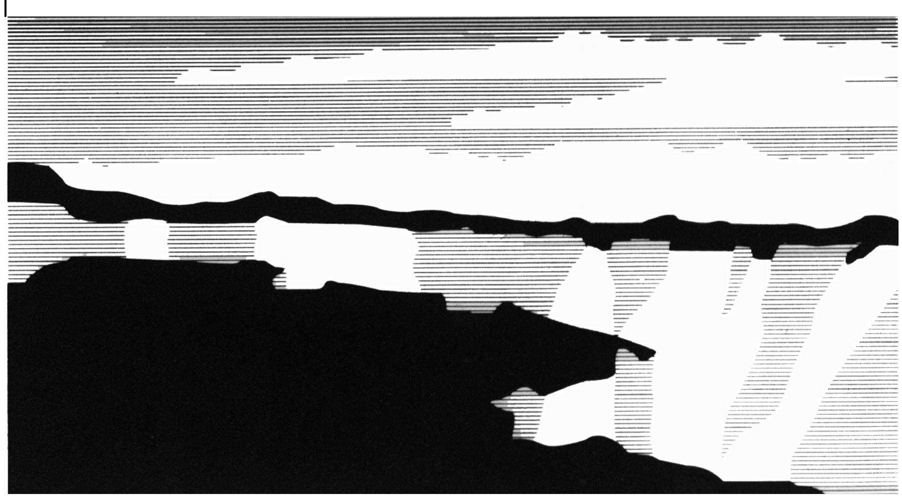

Los Alamos
NATIONAL LABORATORY

Validation of HLW Disposal Cost Savings at Idaho Falls due to the use of TRUEX/SREX

Environmental Technology Cost Savings Analysis Project (ETCAP)
Los Alamos National Laboratory
S.F. DeMuth

March 9, 2000

Abstract

The Innovative Technology Summary Report (ITSR), "TRUEX/SREX Demonstration", claims a potential \$2-B cost savings due to the use of TRUEX/SREX for the disposal of Idaho Falls HLW. A study was conducted and is reported here to provide an independent review of the cost savings reported in the ITSR. It was concluded by way of this independent review that:

(1) TRUEX plus SREX used in conjunction with cesium removal, versus precipitation or freeze crystallization, does have the potential to save \$1-B to \$2-B with a moderate degree of confidence.

(2) TRUEX plus SREX used in conjunction with cesium removal, versus direct immobilization of the existing HLW without separations, does have the potential to save ~\$2-B with a high degree of confidence.

All of the above mentioned technologies (i.e. TRUEX, SREX, cesium removal, precipitation and freeze crystallization) have or will require a significant level of development prior to deployment. Consequently, the "no separations with direct immobilization" can be considered a reasonable baseline.

Introduction

The TRUEX/SREX technologies of interest are actually a suite of three technologies used in series to remove TRUs, strontium, and cesium from the liquid and calcine HLW. The TRUs are removed with the TRUEX solvent extraction process, strontium with the SREX solvent extraction process, and cesium with an ion exchange (IX) resin such as ammonium molybdophosphate (AMP). Removal of these radionuclides converts most of the HLW to a LLW resulting in less expensive disposal; and consequently, all three technologies must be used to achieve the cost savings.

The precipitation process of interest involves neutralization of the acidic waste (existing calcine will be dissolved) by way of hydroxide which forms metal oxides that precipitate. It is reported that this process should preferentially precipitate poly-valent metal ions and leave mono-valent metal ions such as sodium, potassium, and cesium in solution. This process would then require at least one additional step to remove the cesium; however, this would require treating an alkaline solution rather than acidic as is done with TRUEX/SREX/AMP. Alkaline IX resins such as crystalline silico-titanate (CST) are being developed for potential use at Hanford and Savannah River.

Freeze crystallization involves reducing the acidic solution temperature (existing calcine will be dissolved), and hence solubility of the metal nitrate salts (assuming nitric acid is used to dissolve the calcine). The metals at greatest concentration tend to precipitate first, with those at the least concentration remaining in solution. Typically the radionuclides are at far less concentration than species such as sodium, potassium, aluminum, and zirconium. This process is estimated to achieve a separation efficiency of 65% which is considered adequate for the existing liquid tank waste, but would probably not justify dissolution of the calcine without additional separation processes.

Since all of the previously mentioned separation processes have, or will require, significant development before deployment for Idaho Falls specific waste, the obvious alternative to separations is clacination of remaining liquid waste for interim immobilization followed by direct vitrification for permanent immobilization.

Analysis

The basis for the TRUEX/SREX/IX cost savings reported in the ITSR was a comprehensive systems engineering effort, "ICPP Radioactive Liquid and Calcine Waste Technologies Evaluation Final Report and Recommendation" performed at the Idaho Falls site in the early 1990s. The objective of this validation effort was not to evaluate their systems engineering methodology, but rather verify that the ITSR conclusions do actually correspond with those of the Systems Engineering effort, and compare cost savings from other sites using similar technologies. An example of the latter is to compare the cost of cesium separation and HLW glass reported in the Hanford EIS with that of the Idaho Falls Systems Engineering study.

The Idaho Falls Systems Engineering study describes a range of remediation possibilities for the liquid and calcine HLW, which ranges from direct permanent immobilization for disposal without separations, to separation of the majority of radionuclides with TRUEX/SREX/IX, or precipitation/IX, or freeze-crystallization/precipitation/IX. These scenarios are shown in Exhibit 1, where scenario #10 was selected as the system engineering baseline and includes the TRUEX/SREX/IX separations. An alternative for comparison which involves direct final immobilization of the waste without separations is scenario #14. Precipitation with additional cesium removal is defined by scenario #5. Freeze crystallization followed by precipitation with cesium removal is defined by scenario # 1. Scenarios #1, #5, #10, and #14 are used for the cost comparison of the TRUEX/SREX ITSR as shown Exhibit 2.

As shown in Exhibit 3, the life-cycle costs for disposal of HLW with TRUEX/SREX/IX (scenario #10) are equal to ~$2.1-B, while those for direct immobilization without separations (scenario #14) are equal to ~$3.5-B. This yields a cost savings with the use of TRUEX/SREX/IX of ~$1.4-B. The life-cycle costs for disposal of HLW with precipitation/IX (scenario #5) are equal to ~$3.4-B, and those for freeze-crystallization/precipitation/IX (scenario #1) are equal to ~$4.4-B.

TRUEX/SREX/ IX	Precipitation/ IX	Crystallization/ Precipitation/IX	Direct Immobilization
$2.1-B	$3.4-B	$4.4-B	$3.5-B

Table 1. Disposal costs from systems engineering study

As shown in Exhibit 4, the volume of final immobilized HLW with TRUEX/SREX/IX (scenario #10) separations is ~800 m^3, and that without separations (scenario #14) is ~5000 m^3. This results in an 84% volume reduction for final immobilized HLW due to the use of TRUEX/SREX/IX. An 84% volume reduction is reasonable considering the high separation efficiency of these operations as shown by Exhibit 5 and Exhibit 6. Decontamination factors (Dfs) for radionuclides versus nonradionuclides is excellent for the TRUEX process as shown by Exhibit 5, and excellent for AMP IX resin as shown by Exhibit 6 for acid dissolved sludge and acidified supernate which are similar to the liquid and proposed acid dissolved calcine at Idaho Falls. The SREX Dfs derived from the ITSR and shown in Exhibit 4, are marginal at best for potassium and zirconium. However,. However; the HLW at Idaho Falls averages only ~1 wt% potassium and HLW glass is robust for potassium allowing up to 20 wt% potassium plus sodium, and a recent study by Wood in 1997 (see Exhibit 7) indicates SREX should not have a problem with zirconium

Cost estimates for remediation of HLW at the Hanford site can be used to aid in the validation of the Idaho Falls Systems Engineering effort. The estimated Hanford HLW glass based on the original TWRS flow sheet (Orme) was 9300 m^3 as defined by stream #35 of Exhibit 8. The capital plus operating costs for HLW immobilization and HLW disposal at Hanford, based on the TWRS EIS (Slaathaug) were respectively \$2.9-B and \$5.9-B as defined by Exhibit 9. The volumetric cost for HLW immobilization and disposal at Hanford can be estimated as follows.

$$\frac{\$(2.9+5.9)\text{x}10^9}{\left(9300\ \text{m}^3\right)} = \$9.5\text{x}10^5 / \text{m}^3$$

If the cost for HLW immobilization and disposal at Hanford is applied to the HLW volumetric savings at Idaho Falls due to the use of TRUEX/SREX/IX, a cost savings of ~\$4-B is realized.

$$\left(\$9.5\text{x}10^5/\text{m}^3\right)\left(5000\text{m}^3 - 800\text{m}^3\right) \approx \$4\text{x}10^9$$

A similar effort for LLW finds an increased cost at Idaho Falls due to the use of TRUEX/SREX/IX of ~\$0.1B. Additionally, the cesium separation facility with infrastructure at Hanford was estimated as ~\$1.2-B per the TWRS EIS. While the Idaho Falls treatment would include TRUs and strontium in addition to cesium, the amount of liquid waste processed at Hanford has about six times the activity of that processed at Idaho Falls (see Exhibit 10). Taking both of these opposing facts into consideration, an estimate of ~\$1-B for capital and operating expenses for TRUEX/SREX/IX is reasonable. Therefore, using the cost numbers from the Hanford EIS leads to an Idaho Falls cost savings due to the use of TRUEX/SREX/IX of ~\$3-B.

+\$4B(HLW) - \$0.1B(LLW) – \$1B(separation facility) ~ \$3-B

The Hanford based estimate for TRUEX/SREX/IX use at Idaho Falls (~\$3-B) is within a factor of two of the Idaho Falls Systems Engineering effort (~\$1.4-B). This should be considered a validation of the cost savings reported in the ITSR, and does support rounding-off of the \$1.4-B estimate from the systems engineering effort to \$2-B for one significant digit as reported in the ITSR. However, the Idaho Falls Systems Engineering estimate should be considered more accurate than the Hanford based estimate, so as not to claim a \$3-B cost savings.

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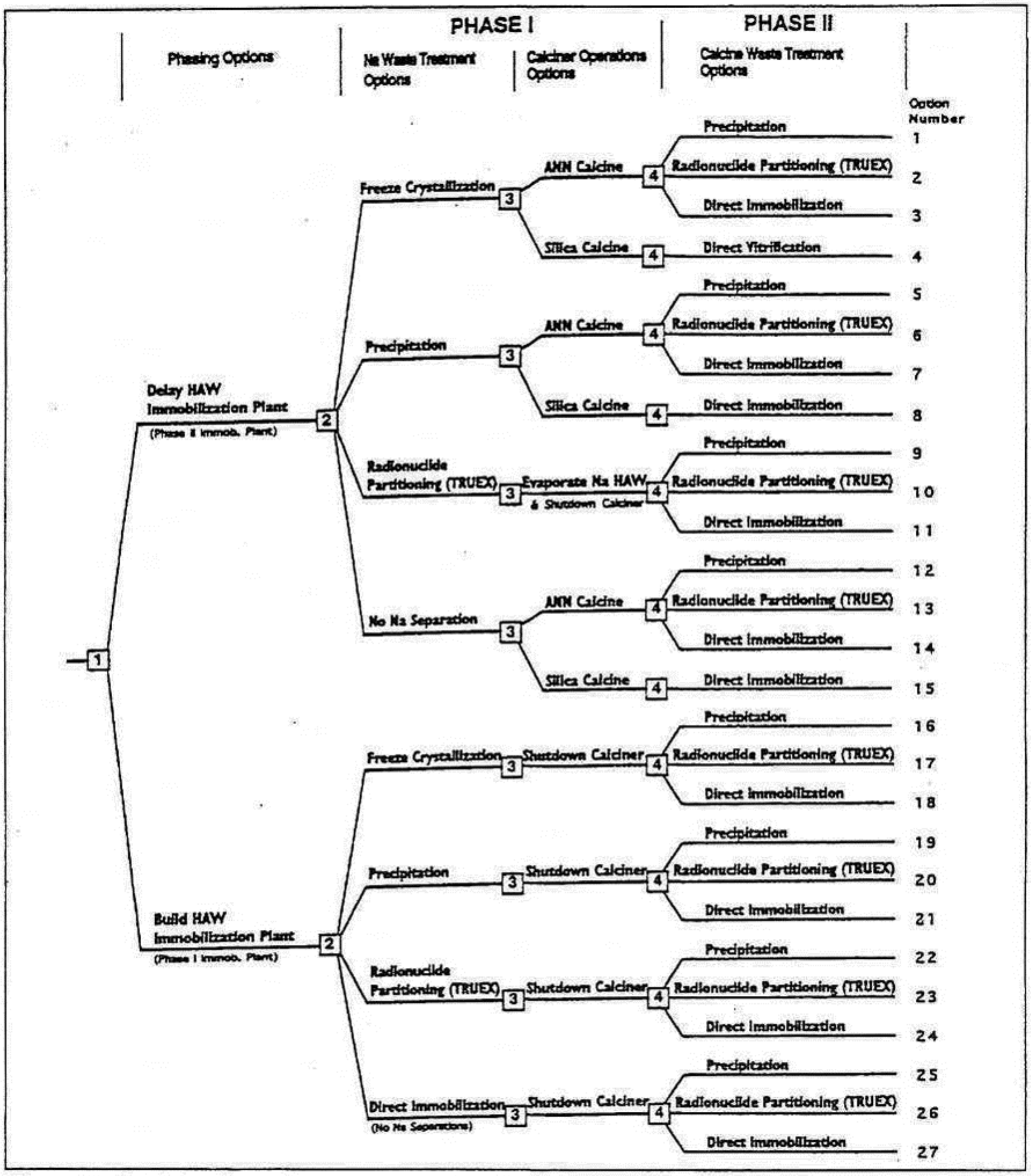

Figure 1: Decision Tree (Nodes 1 through 4) for Waste Treatment Technologies

Exhibit 1. Figure 1 from page 9 of the Systems Engineering study, INEL-94/0119

Table 6. Comparison of TRUEX and SREX to other alternatives (in thousands of dollars)

	TRUEX/SREX	Precipitation and directly immobilized	Freeze crystallization and directly immobilized	No sodium separation and direct immobilized
Development	100	100	100	50
Phase I development & construction costs	500	500	500	200
Phase II development & construction costs	300	1,600	1,600	1,800
Operating costs	900	1,000	1,000	1,000
LLW disposal	200	150	150	50
HLW disposal	200	900	1,100	1,400
D&D of facilities	50	150	150	150
Total estimated cost	2,350	4,400	4,600	4,700
Potential cost savings	N/A	2,050	2,250	2,400

Exhibit 2. Table 6 from page 12 of TRUEX/SREX ITSR, DOE/EM-0419

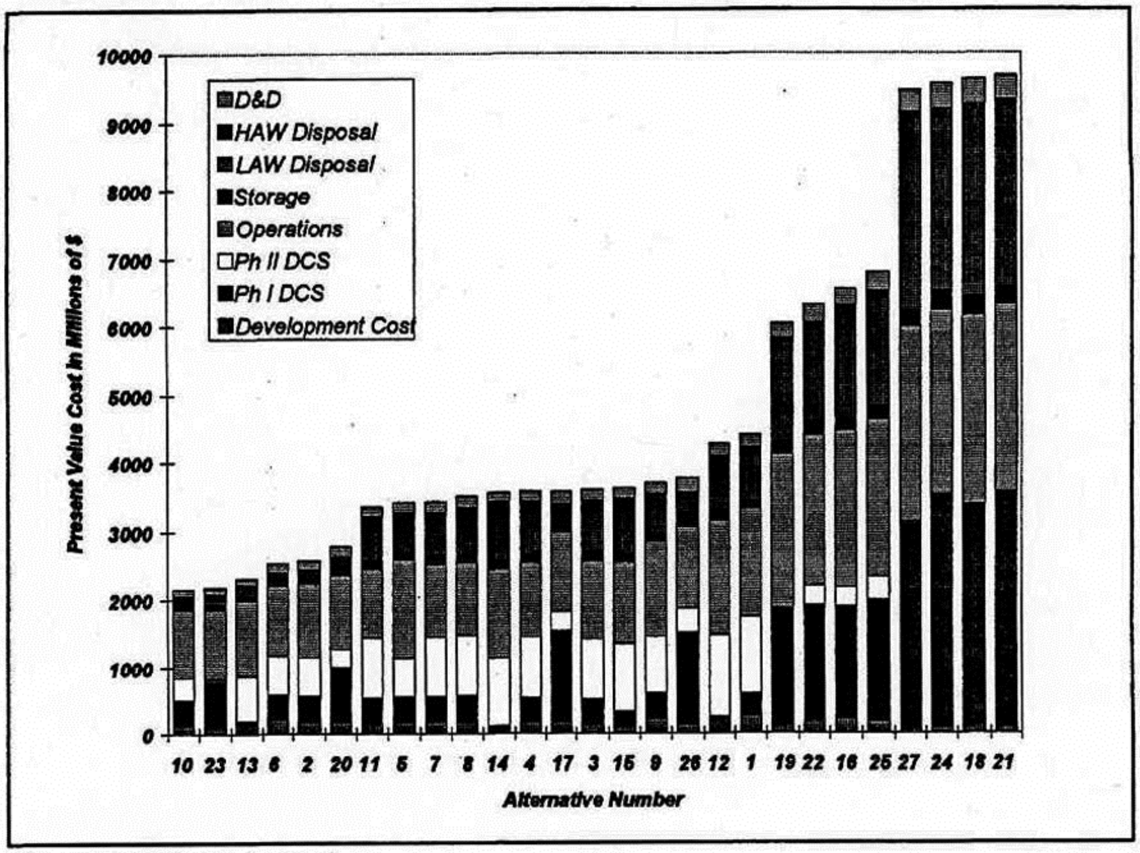

Figure 4: Life-Cycle Costs

Exhibit 3. Figure 4 from page 18 of the Systems Engineering study, INEL-94/0119

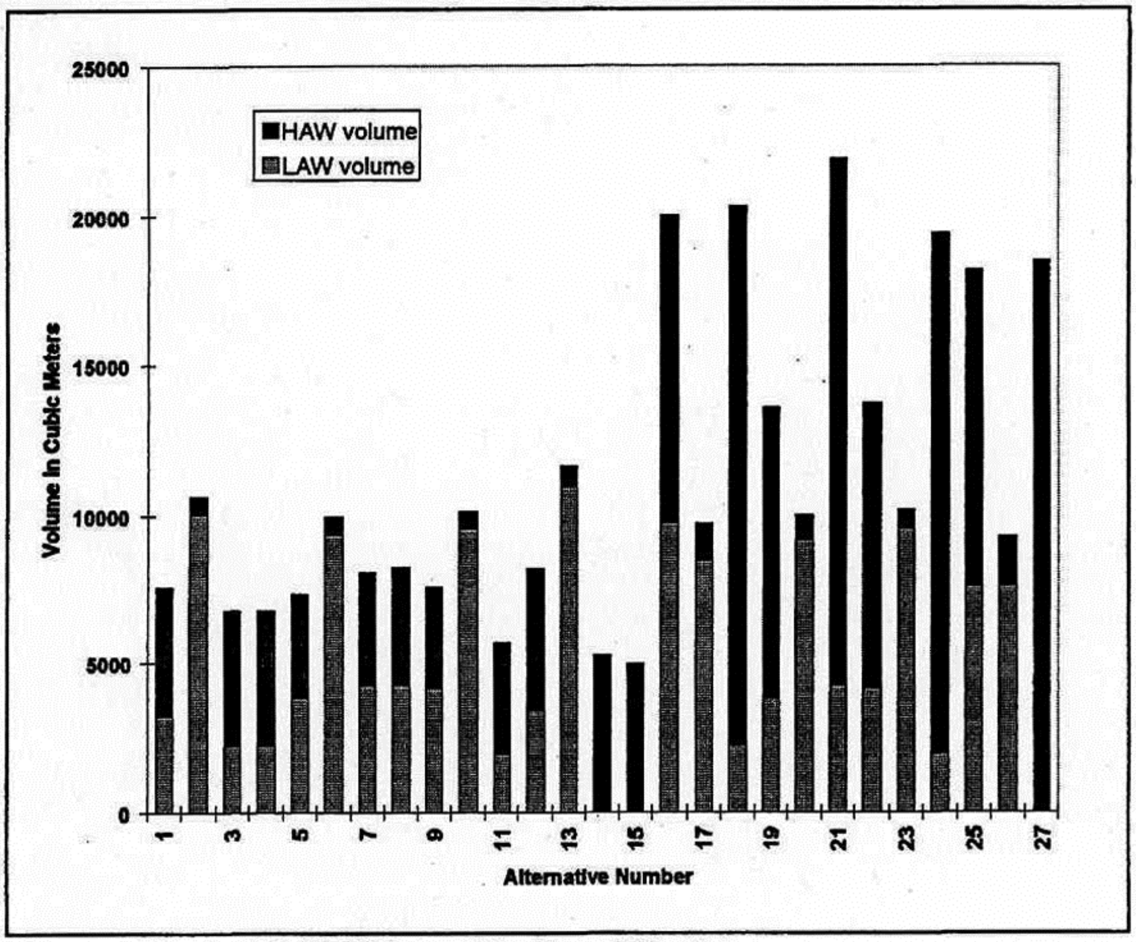

Figure 9: HAW and LAW Volumes After Immobilization

Exhibit 4. Figure 9 from page 23 of the Systems Engineering study, INEL-94/0119

Table 4. Observed removal efficiencies from TRUEX/SREX flow sheet testing

Process	Component	Removal efficiency percentage
TRUEX	Total alpha	99.79
	^{241}Am	99.84
	^{238}Pu	99.97
	^{239}Pu	99.97
	^{235}U	99.85
	^{238}U	99.76
	K	0.06
	Na	0.07
	Fe	0.7
	Hg	74
SREX	^{90}Sr	99.995
	Pb	>95
	Pu	99.94
	U	99.6
	^{241}Am	1.9
	K	37.2
	Hg	89
	Na	0.5
	Zr	81.6
	Ba	64
	Al, B, Cd, Ca, Cr, Cs, Fe, Mn, and Ni are inextractable	

Exhibit 5. Table 4 from page 9 of TRUEX/SREX ITSR, DOE/EM-0419

Table 52. AMP-PAN Composite: Distribution of 14 Elements from Three Simulant Solutions for Hanford Tank 102-SY

Solution	Element	Kd Value for Specified Time		
		30 min	2 h	6 h
Acid-Dissolved Sludge	Ce	0.4	0.2	0.4
	Cs	1626	4072	>5K
	Sr	0.5	0.7	0.9
	Tc	0.8	0.9	1.2
	Y	0.8	0.5	0.4
	Cr	1.0	0.9	1.0
	Co	0.8	0.8	0.8
	Fe	0.1	0.1	0.1
	Mn	0.2	0.1	0.4
	Zn	0.2	0.1	0.2
	Zr	1.4	2.9	4.7
	U	0.2	0.1	0.6
	Pu	0.3	0.4	1.0
	Am	<0.1	<0.1	<0.1
Acidified Supernate	Ce	12	22	28
	Cs	2133	4636	>8K
	Sr	<0.1	<0.1	<0.1
	Tc	0.8	1.2	1.4
	Y	15	27	34
	Cr	0.5	1.3	1.3
	Co	1.5	1.8	1.7
	Fe	0.5	1.1	1.0
	Mn	<0.1	0.1	<0.1
	Zn	0.5	0.4	0.5
	Zr	0.4	1.1	0.9
	U	1.6	2.1	2.5
	Pu	0.5	0.8	1.2
	Am	24	50	86
Alkaline Supernate	Ce	3.4	3.7	4.7
	Cs	0.7	0.4	0.6
	Sr	4.1	2.8	2.6
	Tc	1.4	1.5	1.9
	Y	5.1	5.1	5.2
	Cr	0.5	0.6	0.8
	Co	1.0	0.9	1.0
	Fe	16	17	15
	Mn	3.9	5.5	6.0
	Zn	34	37	29
	Zr	5.2	5.9	6.9
	U	<0.1	1.0	0.8
	Am	1.3	3.2	3.9

Exhibit 6. Ammonium Molybdophosphate (AMP-PAN) K_ds
from page 66 of Marsh, LA-12654

CONCLUSIONS

The evaluation of the SREX Process as a solvent extraction process for the remediation of acidic radioactive liquids at the ICPP has been performed. The laboratory testing (detailed in this report) and counter-current testing (detailed in other reports) indicate that this process is a highly effective method for the removal of ^{90}Sr from tank waste and dissolved calcine wastes at the ICPP.

The decontamination of simulated and actual wastes with respect to ^{90}Sr has been achieved in batch contacts. The decontamination factors obtained in these experiments are suitable high to indicate that the SREX process may be implemented for the treatment of high activity wastes to produce NRC class A low-level wastes.

The use of mixtures of tributyl phosphate and Isopar L® as the diluent for the process solvent has been identified as having physical characteristics which are more desirable than the 1-octanol diluent which was studied in the early phases of this work. It should be noted, however, that the barriers to implementation of a solvent based upon 1-octanol are largely present because of perceived safety hazards and are not viewed as technical problems.

The extractant, DtBuCH18C6, which is commercially manufactured in the United States in multi-kilogram quantities per year, is an effective reagent for the extraction of Sr from the waste streams. The chemical synthesis of this compound is important to its efficiency and wide variation in performance has been observed among manufacturers. This emphasizes the necessity to identify requirements for its performance when large quantities are procured for process-scale operation.

DtBuCH18C6 has been shown to be highly selective for Sr in the presence of many metal ions. The major metals which are extractable by this extractant have been identified as Sr, Pb, Hg, Pu, U, Tc (as TcO_4^-), K, and Ba. Ba is not a major component in ICPP wastes and is unlikely to present obstacles in the working flowsheet. The presence of the other extractables, however, may interfere with the decontamination of the waste stream and may have impacts upon final waste form considerations. The presence of these metals in the aqueous feed to the SREX process need to be carefully addressed in the design of process flowsheets. The stripping of Hg from the loaded solvent has not been fully evaluated. The difficulty of removing the Hg from the SREX solvent must be considered in further development activities.

The radiolysis of the SREX solvent is being studied. Initial results indicate that the radiolytic degradation of the solvent from very low doses of gamma radiation to 1000 KGy have very little effect on its extraction and stripping performance. The distribution coefficients for Sr remain unchanged as a function of absorbed dose in solutions of nitric acid and in simulated waste solutions.

Exhibit 7. SREX conclusions from page 35 of Wood, INEEL/EXT-97-00831

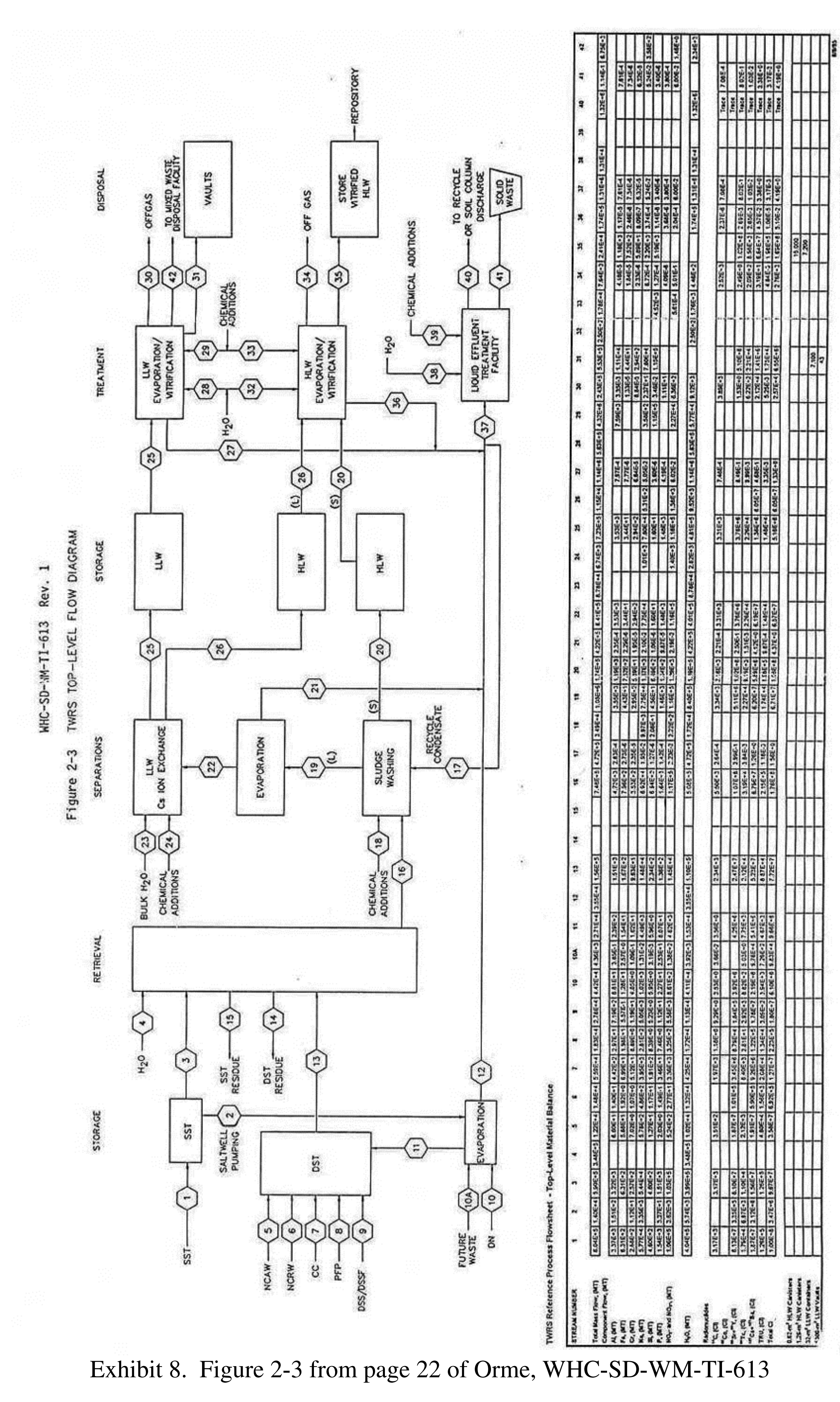

Exhibit 8. Figure 2-3 from page 22 of Orme, WHC-SD-WM-TI-613

	Construction	Labor	Equipment	Materials and Supplies			Low-Level Vaults	HLW Canisters	Research and Development	Repository Fee	Totals
				Start-up	D and D	Operations					
Sludge wash	$43	$129	$6	$2	$12	$6			$9		$207
Cesium removal	$380	$276	$57	$21	$105	$56			$83		~~$975~~ 978
Centralized facilities	$520										$520
LLW vitrification	$1,300	$624	$179	$68	$332	$176			$264		~~$2,934~~ 2943
LLW disposal		$16	$9	$4	$17	$9	$225		$14		$294
HLW vitrification	$1,400	$639	$70	$78	$384	$126			$260		$2,957
HLW transportation		$31									$24
HLW disposal								$239		$5,619	$5,858
Total	$3,643	$1,716	$322	$173	$886	$373	$225	$239	$630	$5,619	$13,826

Notes:

Table values are in millions of 1995 dollars.

Exhibit 9. Table F-36 from page F-93 of Slaathaug, WHC-SD-WM-EV-104

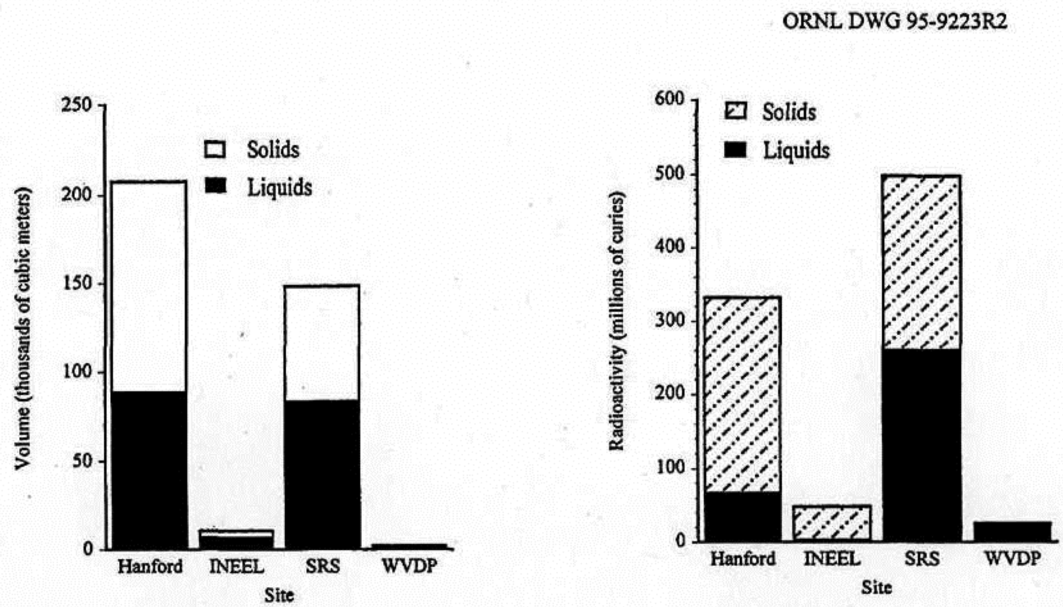

Fig. 2.2. Total volume and radioactivity of solid and liquid HLW through FY 1996.

Exhibit 10. Figure 2.2 from page 69 of the Integrated Data Base Report, DOE/RW-0006, Rev. 12